

## Claims:

1. A membrane electrode unit comprising
  - A) at least one polymer membrane which includes at least one polymer with  
5 at least one nitrogen atom, the polymer membrane including at least one mineral acid,
  - B) at least two electrodes,  
characterized in that at least one electrode includes a catalyst containing
    - i. at least one precious metal of the platinum group, in particular Pt, Pd, Ir,  
10 Rh, Os, Ru, and/or at least one precious metal Au and/or Ag
    - ii. at least one metal less precious according to the electrochemical series as the metal mentioned in (i.), in particular selected from the group of Fe, Co, Ni, Cr, Mn, Zr, Ti, Ga, V.
- 15 2. The membrane electrode unit according to claim 1, characterized in that a polyphosphazene is employed as the polymer with at least one nitrogen atom.
3. The membrane electrode unit according to claim 1, characterized in that an alkaline polymer with at least one nitrogen atom in a repeating unit is used as  
20 the polymer with at least one nitrogen atom.
4. The membrane electrode unit according to claim 3, characterized in that the alkaline polymer contains at least one aromatic ring with at least one nitrogen atom.  
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5. The membrane electrode unit according to claim 4, characterized in that the alkaline polymer is a polyimidazole, a polybenzimidazole, a polybenzothiazole, a polybenzoxazole, a polytriazole, a polyoxadiazole, a polythiadiazole, a polypyrazole, a polyquinoxaline, a poly(pyridine), a poly(pyrimidine) or a  
30 poly(tetrazapyrene).
6. The membrane electrode unit according to claim 3, characterized in that a mixture of one or more alkaline polymers with another polymer is employed.
- 35 7. The membrane electrode unit according to claim 1, characterized in that the membrane comprises phosphoric acid and/or sulphuric acid as the mineral acid, preferably doped in the polymer membrane.

8. The membrane electrode unit according to one or more of claims 1 to 7, characterized in that the polymer membrane comprises para-polybenzimidazoles.
- 5 9. The membrane electrode unit according to one or more of claims 1 to 8, characterized in that the polymer membrane can be obtained by a process comprising the steps of
- 10 i) preparation of a mixture comprising polyphosphoric acid, at least one polyazole and/or at least one or more compounds which are suitable for the formation of polyazoles with action of heat in accordance with step ii),
  - 15 ii) heating the mixture obtainable in accordance with step i) under inert gas to temperatures of up to 400°C,
  - iii) applying a layer using the mixture in accordance with step i) and/or ii) to a support,
  - iv) treatment of the membrane formed in step iii).
- 20 10. The membrane electrode unit according to claim 9, characterized in that the mixture produced in step i) comprises one or more aromatic and/or heteroaromatic tetraamino compounds and one or more aromatic and/or heteroaromatic carboxylic acids or their derivatives, which comprise at least two acid groups per carboxylic acid monomer, and/or one or more aromatic and/or heteroaromatic diaminocarboxylic acids which are suitable for the formation of
- 25 polyazoles with action of heat in accordance with step ii).
- 30 11. The membrane electrode unit according to claim 9, characterized in that the mixture produced in step i) comprises compounds which are obtainable by reaction of one or more aromatic and/or heteroaromatic tetraamino compounds with one or more aromatic and/or heteroaromatic carboxylic acids or their derivatives, which contain at least two acid groups per carboxylic acid monomer, or of one or more aromatic and/or heteroaromatic diaminocarboxylic acids in the melt at temperatures of up to 400°C, which are suitable for the formation of polyazoles with action of heat in accordance with step ii).
- 35 12. The membrane electrode unit according to claim 10 or 11, characterized in that the compounds suitable for the formation of polyazoles as aromatic and/or heteroaromatic tetraamino compound comprise compounds which are selected

from the group consisting of 3,3',4,4'-tetraaminobiphenyl, 2,3,5,6-tetraaminopyridine and/or 1,2,4,5-tetraaminobenzene.

13. The membrane electrode unit according to claim 10, 11 or 12, characterized in that the compounds suitable for the formation of polyazoles as aromatic and/or heteroaromatic carboxylic acids or their derivatives, which contain at least two acid groups per carboxylic acid monomer, comprise compounds which are selected from the group consisting of isophthalic acid, terephthalic acid, phthalic acid, 5-hydroxyisophthalic acid, 4-hydroxyisophthalic acid, 2-hydroxyterephthalic acid, 5-aminoisophthalic acid, 5-N,N-dimethylaminoisophthalic acid, 5-N,N-diethylaminoisophthalic acid, 2,5-dihydroxyterephthalic acid, 2,5-dihydroxyisophthalic acid, 2,3-dihydroxyisophthalic acid, 2,3-dihydroxyphthalic acid, 2,4-dihydroxyphthalic acid, 3,4-dihydroxyphthalic acid, 3-fluorophthalic acid, 5-fluoroisophthalic acid, 2-fluoroterephthalic acid, tetrafluorophthalic acid, tetrafluoroisophthalic acid, tetrafluoroterephthalic acid, 1,4-naphthalenedicarboxylic acid, 1,5-naphthalenedicarboxylic acid, 2,6-naphthalenedicarboxylic acid, 2,7-naphthalenedicarboxylic acid, diphenic acid, 1,8-dihydroxynaphthalene-3,6-dicarboxylic acid, diphenyl ether-4,4'-dicarboxylic acid, benzophenone-4,4'-dicarboxylic acid, diphenylsulphone-4,4'-dicarboxylic acid, biphenyl-4,4'-dicarboxylic acid, 4-trifluoromethylphthalic acid, 2,2-bis-(4-carboxyphenyl)hexafluoropropane, 4,4'-stilbenedicarboxylic acid, 4-carboxycinnamic acid or their C1-C20 alkyl esters or C5-C12 aryl esters or their acid anhydrides or their acid chlorides.
14. The membrane electrode unit according to claim 10, 11, 12 or 13, characterized in that the compounds suitable for the formation of polyazoles comprise aromatic tricarboxylic acids, their C1-C20 alkyl esters or C5-C12 aryl esters or their acid anhydrides or their acid halides, or tetracarboxylic acids, their C1-C20 alkyl esters or C5-C12 aryl esters or their acid anhydrides or their acid halides.
15. The membrane electrode unit according to claim 14, characterized in that the aromatic tricarboxylic acids comprise compounds which are selected from the group consisting of 1,3,5-benzenetricarboxylic acid (trimesic acid); 2,4,5-benzenetricarboxylic acid (trimellitic acid); (2-carboxyphenyl)iminodiacetic acid, 3,5,3'-biphenyltricarboxylic acid; 3,5,4'-biphenyltricarboxylic acid; 2,4,6-pyridinetricarboxylic acid, benzene-1,2,4,5-tetracarboxylic acids; naphthalene-1,4,5,8-tetracarboxylic acids, 3,5,3',5'-biphenyltetracarboxylic

acids, benzophenonetetracarboxylic acid, 3,3',4,4'-biphenyltetracarboxylic acid, 2,2',3,3'-biphenyltetracarboxylic acid, 1,2,5,6-naphthalenetetracarboxylic acid and/or 1,4,5,8-naphthalenetetracarboxylic acid.

5 16. The membrane electrode unit according to claim 14 or 15, characterized in that the content of tricarboxylic acid and/or tetracarboxylic acids is between 0 and 300 mol-%, preferably 0.1 and 20 mol-%, in particular 0.5 and 10 mol-%, based on dicarboxylic acid used.

10 17. The membrane electrode unit according to one or more of claims 10 to 16, characterized in that the compounds suitable for the formation of polyazoles comprise heteroaromatic dicarboxylic acids, tricarboxylic acids and/or tetracarboxylic acids, which contain at least one nitrogen, oxygen, sulphur or phosphorus atom in the aromatic group.

15 18. The membrane electrode unit according to claim 17, characterized in that pyridine-2,5-dicarboxylic acid, pyridine-3,5-dicarboxylic acid, pyridine-2,6-dicarboxylic acid, pyridine-2,4-dicarboxylic acid, 4-phenyl-2,5-pyridinedicarboxylic acid, 3,5-pyrazoledicarboxylic acid, 2,6-  
20 pyrimidinedicarboxylic acid, 2,5-pyrazinedicarboxylic acid, 2,4,6-pyridinetricarboxylic acid, benzimidazole-5,6-dicarboxylic acid and their C1-C20 alkyl esters or C5-C12 aryl esters or their acid anhydrides or their acid chlorides are employed.

25 19. The membrane electrode unit according to claim 10 or 11, characterized in that the compounds suitable for the formation of polyazoles comprise diaminobenzoic acid and/or its monohydrochloride and dihydrochloride derivatives.

30 20. The membrane electrode unit according to any one of claims 10 to 19, characterized in that at least one para-dicarboxylic acid is employed.

35 21. The membrane electrode unit according to one or more of claims 10 to 20, characterized in that the heating in accordance with step ii) is performed after the formation of a flat structure in accordance with step iii).

22. The membrane electrode unit according to any one of claims 10 to 21, characterized in that the membrane formed according to step iv) has a thickness between 15 and 3000  $\mu\text{m}$ .

5 23. The membrane electrode unit according to any one of claims 1 to 22, characterized in that the catalyst contains

- i. at least one precious metal of the platinum group, in particular Pt, Pd, Ir, Rh, Os, Ru, and/or at least one precious metal Au and/or Ag
- 10 ii. at least one metal less precious according to the electrochemical series as the metal mentioned in (i.), in particular selected from the group of Fe, Co, Ni, Cr, Mn, Zr, Ti, Ga, V.

24. The membrane electrode unit according to any one of claims 1 to 23, characterized in that the catalyst is applied to the polymer membrane.

15 25. The membrane electrode unit according to one or more of claims 1 to 24, characterized in that the catalyst layer has a thickness in the range of from 0.1 to 50  $\mu\text{m}$ .

20 26. The membrane electrode unit according to one or more of claims 1 to 25, characterized in that the catalyst comprises catalytically active particles which have a size in the range of from 5 to 200 nm.

25 27. The membrane electrode unit according to one or more of claims 1 to 26, characterized in that the catalyst comprises catalytically active particles on a support, the size of the catalyst particles being in the range of from 1 to 20 nm.

30 28. The membrane electrode unit according to one or more of claims 1 to 27, characterized in that the membrane electrode unit comprises 0.01 to 20  $\text{g}/\text{m}^2$ , preferably 0.1 to 10  $\text{g}/\text{m}^2$ , of a catalytically active substance.

29. The membrane electrode unit according to claim 27, characterized in that the catalytically active particles include carbon as a support.

35 30. The membrane electrode unit according to one or more of claims 1 to 29, characterized in that the weight ratio of the precious metals of the platinum group or of Au and/or Ag (i) to the metals less precious according to the electrochemical series (ii) is between 1:100 to 100:1.

31. A fuel cell containing one or more membrane electrode units according to one or more of claims 1 to 30.